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**14. ABSTRACT**
Our primary research effort during the funding period focused on the exploration and control of coherent time-dependent electronic interactions within and between atoms. Specifically, we considered the behavior of Rydberg atoms in which one electron is very highly-excited, residing in a state of large principal quantum number, n. Such highly-excited Rydberg electrons are extremely sensitive to electric fields due, for example, to neighboring atoms or external radiation at frequencies from DC to THz. The strong coupling between cold Rydberg atoms is at the core of some proposed quantum logic implementations which seek to control entangling interactions between atoms. Our recent experiments have verified the controllability of coherent long-range few-body interactions and demonstrated electronic coherence times which are comparable to the time-scales for relative atom motion and the spontaneous emission lifetime of the Rydberg states. In addition, we have developed an ultra-fast, potentially single-shot detector of THz waveforms. The detector exploits the large polarizability and electric field sensitivity of Rydberg atoms to enable the visualization of THz waveforms with sub-picosecond resolution over 100 picosecond intervals.

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## Abstract

During the funding period, our primary research efforts have focused on the exploration and control of coherent time-dependent electronic interactions within and between atoms. Specifically, we have examined the response of ensembles of Rydberg atoms to each other and to externally applied THz radiation. By definition, one or more electrons in a Rydberg atom are highly excited, and reside in a state of large principal quantum number, n. Such electrons are extremely sensitive to electric fields due, for example, to neighboring atoms or external radiation at frequencies from DC to THz. The strong, long-range coupling between Rydberg atoms is at the core of some proposed quantum logic implementations, but these require control of coherent, entangling interactions between atoms. Our recent experiments have verified the controllability of resonant few-body dipole-dipole interactions in cold gases and demonstrated electronic coherence times which are comparable to the time-scales for relative atom motion and the spontaneous emission lifetime of the Rydberg states. These coherence times are one to two orders of magnitude longer than those suggested by previous measurements, improving the likelihood of successful quantum logic implementations in these systems. In addition, we have developed an ultra-fast, potentially single-shot detector of THz waveforms. The detector exploits the large polarizability and electric field sensitivity of Rydberg atoms and could enable the visualization of THz waveforms with sub-picosecond resolution over 100 picosecond intervals.

## I. Introduction

For the most part, research in atomic physics has moved beyond passive spectroscopy of isolated atoms. New laser and electro-optic technology and techniques serve as the key tools for active manipulation of inter- and intra-atomic dynamics. Indeed, optical control methods are enabling entirely new classes of atomic physics research, with fundamental science and practical applications from designer many-body systems that simulate model condensed matter Hamiltonians, to quantum information storage and processing, to new radiation sources and detectors.

The experiments performed during this AFOSR funding period were designed to exploit the extreme electric field sensitivity of Rydberg atoms. In contrast to ground- or low-lying excited-state atoms, the application of resonant or non-resonant, time-dependent electric fields of a few to a few thousand V/cm can severely alter the electronic eigenstates within Rydberg atoms. For example, in one line of experiments, angular momentum mixing in Rydberg atoms exposed to THz radiation was used to enable a novel THz waveform detector.

More generally, the application of the appropriate fields can be used to coherently manipulate Rydberg atoms, essentially transforming the quantum state at will. Such transformations require that coherence between the relevant, field-dressed, atomic states be maintained. This is readily accomplished with isolated atoms, where the atom-field coupling can be made much larger than decoherence rates due, for example, to stray fields, collisions with background gas, or spontaneous emission. In that case, the desired transformations can be accomplished in femtoseconds to nanoseconds, well within the microsecond to millisecond coherence times of the system. Moreover, as we recently showed, time-varying electric fields and pulse sequences can be used to actively stabilize isolated Rydberg atoms, essentially immunizing them from electronic decoherence.

However, in the presence of atom-atom interactions the situation is considerably more complex as the MHz-scale strength of couplings between Rydberg atoms can be comparable to decoherence rates. Moreover, in an ensemble of interacting atoms, the border between the "system" and its "environment" may not be so well defined. Thus, in a second line of experiments, we sought to determine whether dipole-dipole interactions between Rydberg atoms could be coherently manipulated over extended time periods. Accordingly, we developed a novel coherent spectroscopy to measure coherence times in the presence of dipole-dipole interactions in a cold Rydberg gas in a magneto-optical trap. We also demonstrated a novel technique for reducing the relative velocities of neighboring atoms in an atomic beam to enable the exploration of coherent interactions between atoms and/or molecules that are not easily laser cooled or optically trapped.

## II. Results

The results of our AFOSR sponsored research are described in four articles and in the Ph.D. dissertation of Dr. Mary Kutteruf, all of which were published during the grant period (see list in Sect. III). Two additional manuscripts are nearly ready for submission to Physical Review Letters and Optics Express, respectively. The principal results from the current funding period are described below.

### *A. An Atomic Oscilloscope for Viewing THz Waveforms*

We have demonstrated a THz bandwidth oscilloscope for direct measurement of the time-dependent electric field of freely propagating THz waveforms. The technique exploits the dynamical Stark

effect in low-lying atomic Rydberg states to enable non-invasive, self-calibrating field detection through photo-excitation of dipole forbidden transitions. The instrument utilizes counter-propagating THz and laser beams and spatial resolved photoexcitation detection. It is similar in concept to electro-optic sampling (EOS) via optical polarization gating in a non-linear crystal, but is non-invasive, as it is based on THz gating of optical excitations in a low density atomic gas. The method allows for precise THz electric field calibration using a static electric field reference. Moreover, because the physical detector is a low density gas, it is non-dispersive, and can be used to measure THz fields *in situ*, within devices or experimental apparatus.

To demonstrate the method, we use two laser pulses with durations of 5 ns and 800 fs, to photo-excite Li atoms from the 2s ground state, through the 2p level, to n=6 Rydberg states, respectively. The excitation proceeds in the presence of both DC and THz electric fields. A third, 120 fs, laser pulse photoionizes any Rydberg population produced by the first two laser pulses. Information regarding the THz field is obtained from the influence that net electric field has on the Rydberg excitation and subsequent ionization probability as a function of the delay between the Rydberg excitation pulse and the THz waveform.

To demonstrate the method we measured the time-dependent THz field generated from a GaAs photoconductive switch that was triggered by a 120 fs laser pulse. To perform the measurement, the laser-THz-atom interaction region were positioned between parallel electric field plates. Photo-ions produced during the interaction of the atoms with the lasers and THz radiation were pushed through a slit in the upper field plate toward a spatially sensitive MCP-Phosphor detector combination. The counter propagation of the THz and laser beams enabled the mapping of ionization probability, as a function of the delay $t_0$ between the Rydberg excitation-THz pulse, onto the spatial axis defined by the slit. By recording the position dependent ionization signal using a CCD camera, we can obtain the delay-dependent excitation signal in a single laser shot.

In the absence of an external electric field, the single-photon excitation 2p → 6p is dipole forbidden. However, in the presence of an electric field F, the nominal 6p state acquires character of other angular momentum states through the Stark effect. In particular, for field magnitudes F < 10 kV/cm, the nominal |6p> level undergoes only a small second order Stark shift, but the state vector transforms into $\left|\psi_{6p}\right\rangle = \left|6p\right\rangle + \frac{Fz_0}{\Delta E}\left|6d\right\rangle$, where $z_0 = \langle 6d|z|6p\rangle$ and ΔE is the zero-field energy splitting between the 6p and 6d levels. Dipole excitation of $|\psi_{6p}\rangle$ from |2p> proceeds through the Stark induced |6d> character. Therefore, when the Rydberg laser is tuned to $|\psi_{6p}\rangle$, the excitation probability is proportional to the net electric field $F(t)^2 = (F_{DC} + \langle F_{THz}(t)\rangle)^2$ where $\langle F_{THz}(t)\rangle$ is the average THz field during the Rydberg excitation pulse. For a sufficiently short Rydberg excitation pulse, $\langle F_{THz}(t)\rangle = F_{THz}(t)$ and sufficiently weak fields $F_{THz}(t) \ll F_{DC}$, at a given delay, t, the difference in the ionization probabilities, with and without the THz field present, normalized by the signal without the THz field is $(P_{THz}-P_{DC}) / P_{DC} = 2\, F_{THz}(t)$.

Straight forward improvements to the current device could enable single-shot viewing of freely propagating waveforms with ~100 femtosecond resolution over time intervals exceeding 100 picoseconds. The technique might be extended beyond THz waveform analysis to applications such as 3-dimensional THz imaging. A manuscript describing this work is nearly ready for submission to Optics Express

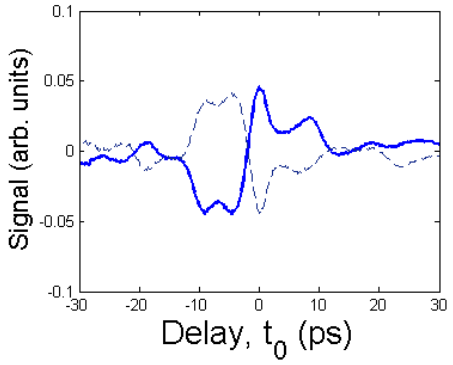


**Figure 1:** Difference in excitation probabilities ($P_{THz}$-$P_{DC}$), with and without the THz field present, for positive and negative polarity THz fields as recorded with CCD camera using the single-shot, counter-propagating pump-probe geometry. The time-dependent signal is proportional to the time-dependent electric field in the THz pulse averaged over the 800 fs duration of the Rydberg excitation laser and the spatial resolution of the CCD camera.

*B. Rydberg Atom Coherence in the Presence of Dipole-Dipole Interactions in a Cold Gas*

Proposals to implement quantum logic operations via controlled dipole-dipole interactions between Rydberg atoms have generated considerable interest in the coherent manipulation of these systems and in understanding coherence within them. Measurements made in several different groups have shown rapid dephasing (< 1μs) of Rydberg excitations in the presence of dipole-dipole interactions in cold Rydberg gases. During the funding period we developed a novel coherent spectroscopy that enables us to extract microscopic coherence rather than macroscopic dephasing rates for these systems. Using this method we have found that microscopic coherence times in cold Rydberg gases can exceed 10μs and likely are limited primarily by residual atom motion.

Consider two, isolated, 2-level Rydberg atoms, A and B, with opposite parity eigenstates |S>, |P> and |S'>, |P'>, and dipole moments $\mu \sim \langle S|z|P\rangle$ and $\mu' \sim \langle S'|z|P'\rangle$, respectively. Assume that the S-P energy splitting in A is nearly the negative of that in B such that $(E_S + E_{S'}) \approx (E_P + E_{P'}) \approx E_0$, and $(E_S + E_{S'}) - (E_P + E_{P'}) = \delta$. Given the high density of Rydberg states, it is straightforward in practice to identify suitable states, since δ can be Stark tuned through the application of static or pulsed electric fields. When separated by a distance R, atoms A and B are coupled by a dipole-dipole interaction $V_0 = \mu_A\mu_B/R^3$. Due to the coupling, the atom pair has two eigenstates which are nearly degenerate, with energies $E_+$ , $E_- \approx E_0$. These eigenstates are entangled superpositions of the eigenstates of the isolated atoms

$$|+\rangle = \cos\theta\, |S\rangle|S'\rangle + \sin\theta\, |P\rangle|P'\rangle$$
$$|-\rangle = -\sin\theta\, |S\rangle|S'\rangle + \cos\theta\, |P\rangle|P'\rangle.$$

Here θ is an admixture parameter that depends on the ratio of $V_0$ to the energy splitting δ of the uncoupled atom pairs ( $\theta \rightarrow 0,\pi/2$ for $|\delta| \gg V_0$ and $\theta \rightarrow \pi/4$ for $\delta \ll V_0$).

We assume that at t=0 the two atoms are in a low lying P-state (e.g. the upper trap state in a MOT) when they are exposed to two short laser pulses which are tuned to excite the individual atoms to states |S> and |S'>, respectively. The lasers create a coherent superposition of the Rydberg pair states |+> and |->. If the coherent bandwidths of the lasers are much greater than $V_0$ and |δ|, and all other interactions can be ignored, then at t=0, the wavefunction for the Rydberg pair is Ψ(0) = |S>|S'>. Assuming coherence is maintained, after the excitation lasers, the wavefunction for the atom pair evolves in the presence of the dipole-dipole interaction according to the two level Rabi formula:

$$\Psi(t) = (\cos\phi - i\eta \sin\phi)\, |S\rangle|S'\rangle + i\xi \sin\phi\, |P\rangle|P'\rangle$$

where $\phi = \gamma t/2$ is the Rabi phase, $\gamma = \sqrt{4V_0^2 + \delta^2}$ is the Rabi frequency, $\xi = 2V_0/\gamma$, and $\eta = \delta/\gamma$. Consequently, the probability of finding atoms in |P> and |P'> following the laser excitation is $\mathcal{P} = \xi^2 \sin^2(\gamma t/2)$. Thus $\mathcal{P}$ exhibits Rabi oscillations with amplitude $\xi^2$ and frequency $\gamma$.

In an experiment in which an ensemble of atoms is excited and detected, the variation in interatomic separations, R, results in a distribution of Rabi frequencies and oscillation amplitudes over the ensemble. As a result, a monotonic increase and eventual saturation, but no Rabi oscillations, are observed in the p-state population as a function of delay following the laser excitation. The lack of Rabi oscillations does not imply microscopic decoherence among individual atom pairs, but instead is a reflection of macroscopic inhomogeneities in the ensemble.

Of course, an additional complication when dealing with an ensemble is that each pair of atoms, A and B, is also influenced by other atoms. Additional interactions, mediating exchange or "hopping" process such as $|S\rangle|S\rangle|P\rangle \rightarrow |S\rangle|P\rangle|S\rangle$, $|P\rangle|P'\rangle|S\rangle \rightarrow |S\rangle|P'\rangle|P\rangle$, and myriad other combinations involving three or more atoms are also at play. Accordingly, the nearly degenerate many-body eigenstates no longer have the simple form of the |+> and |-> pairs defined above. However, if one prepares the system using short laser pulses such that only atoms in states |S> and |S'> are initially excited, the system should still evolve coherently into |P> and |P'> at some rate and with some maximum probability, due to the mutual interactions between atoms. The question is: how does one probe or confirm the coherence of this few- or many-body process in the presence of inhomogenieties associated with the distribution of atom separations in an ensemble?

In our experiments, we use 5 ns pulsed dye lasers to excite cold ( ~70μK) Rb atoms in a MOT to |25s> and |33s> Rydberg levels. Using state-selective field ionization, we detect population transfer, via dipole-dipole interactions, to the $|24p_{1/2}\rangle$ and $|34p_{3/2}\rangle$ levels. Working with this identical system, Tom Gallagher's group at Virginia previously measured the dephasing rate of the dipole-dipole mediated process. Using a variant of Ramsey's separated oscillatory fields method they allowed atoms to interact on-resonance (i.e. $\delta=0$) for a short time $t_0$ at $\delta=0$, switched off the interaction between atoms for a time T using an electric field pulse to detune them off resonance (i.e. $\delta >> V_0$), and then allowed them to interact on-resonance for an additional time $t_0$. They observed oscillations in the measured p-state signal due to the interference between the probability amplitudes transferred during the initial and final, on-resonance, interaction times. The oscillations decayed as the delay T between the two interaction times was increased, suggesting a dephasing time of ~ 100 ns at Rydberg atom densities of ~ $10^9$ $cm^{-3}$. They attributed the rapid dephasing to the beyond nearest neighbor "hopping" or exchange processes described above. We have used a different pulsed field approach that has enabled us to show that the microscopic coherence times in this system can be two orders of magnitude longer than this dephasing time.

In our method, atoms are initially excited with a detuning $\delta = +\delta_0$ and allowed to interact for a time T/2. A pulsed voltage is then used to suddenly change the detuning to $\delta = -\delta_0$, and the atoms are allowed to interact for an additional time T/2 before the net p-state population is measured. The signal acquired using the tuning "jump" is compared to that obtained with "no jump", i.e. with the atoms held at constant detuning, $\delta = +\delta_0$ or $\delta = -\delta_0$, for the full time interval, T. For two atoms, it is straightforward to show that due to interference between the two distinct interaction times, as long as coherence is maintained, the probability for finding the atoms in a p-state following the jump is always greater than that without a jump, *regardless of the magnitude of $\delta_0$ or the internuclear*

*separation, R*. Our numerical simulations using 3 atoms confirm that this coherent enhancement persists in the presence of beyond nearest neighbor interactions as well. In addition, both the two atom model and the 3-body simulation predict an increase in the coherent signal enhancement as the number of detuning jumps is increased. Conversely, since the p-state excitation probabilities are identical for $\delta = +\delta_0$ or $\delta = -\delta_0$, *if coherence has been lost during the first T/2 interaction interval*, the excitation probability obtained with a detuning jump will be identical to that measured (for the same total time interval, T) with no jump. Thus, by measuring the decrease in the p-state signal enhancement for one or more detuning jumps, we can determine the microscopic decoherence rate in the ensemble.

Figure 2 shows the measured p-state signal for a Rydberg atom density of ~2 x $10^9$ $cm^{-3}$ as a function of total time interaction time for 0, 1, 3, 7, and 15 detuning jumps. Also shown in the figure are the associated signal enhancement curves as a function of total interaction time, and the decoherence rates derived from those curves. The measured signal enhancement curves indicate coherence times on the order of 10 μs, two orders of magnitude longer than the dephasing times measured previously.

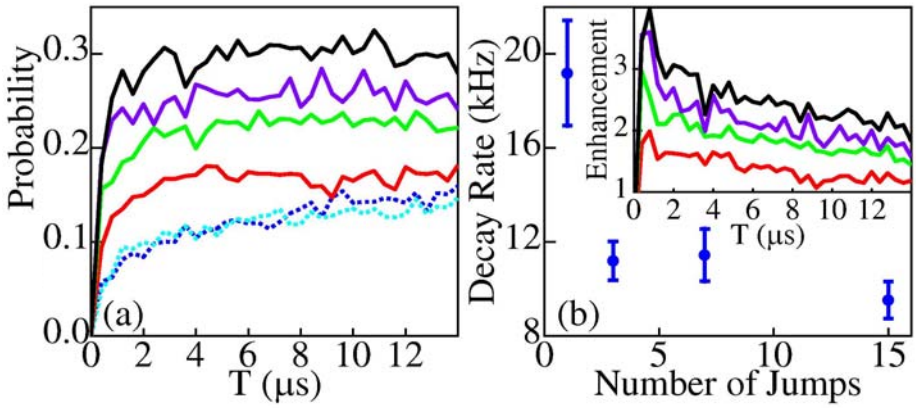


**Figure 2:** (Left) Measured p-state signal as a function of the total interaction time T for 1 (red), 3 (green), 7 (purple) and 15 (black ) detuning jumps. The dashed blue and dashed cyan lines are the measured signals acquired for constant detunings $\delta_0 = \pm 5.5$ MHz, respectively. (Right) The signal enhancement decay rates vs. the number of detuning jumps obtained from the delay-dependent enhancement signal shown in the insert.

Numerical simulations that crudely model the average motion of atoms in the MOT indicate that atom motion and the associated time-dependent changes in the dipole-dipole couplings should result in decoherence over this same 10 μs time-scale. Interestingly, there is some indication in the data that more frequent jumps can extend the coherence times. This is reminiscent of the decoherence suppression that we observed in fine-structure wavepackets following the application of multiple system kicks which periodically flipped the state vector, erasing the slow acquisition of environmental noise. Additional work will be required to determine i) if multiple jumps actually result in a reduction of decoherence in this system, and ii) the specific physical mechanism responsible for that reduction. A manuscript describing this work is nearly ready for submission to Physical Review Letters.

*C. Controlled Dipole-Dipole Interactions Between Rydberg Atoms in a Laser-Chopped Effusive Beam*

We have demonstrated a laser-based atomic beam chopper to explore coherent interactions between Rydberg atoms without the need for laser cooling or trapping. Specifically, free expansion of a localized, laser-excited sample of Rydberg atoms is used to reduce the relative velocities of neighboring Rydberg atoms prior to the initiation of Rydberg-Rydberg interactions between them. Specifically, we have used this laser-chopper technique to study the resonant energy exchange process, $|29s\rangle|27d_{3/2}\rangle \rightarrow |29p_{1/2}\rangle |28p_{1/2}\rangle$, that is mediated by a dipole-dipole coupling between K atoms. In the experiments, pulsed electric fields exploit the Stark effect to tune pairs of atoms into and out of resonance, effectively gating the resonant interaction. We have demonstrated that the relative atom velocities can be reduced to the point where the duration of the electric field tuning pulses, not the motion of neighboring atoms, defines the interaction time for each atom pair within the ensemble. We have observed coherent, transform-limited broadening of the resonance lineshapes as the tuning pulse duration is reduced below the natural time-scale for collisions. This technique, alone or in combination with supersonic expansion, may make it possible to study coherent interactions between atoms or molecules which are not easily laser cooled or trapped. A manuscript describing this work was recently published in Physical Review A.

## III. Publications Resulting from AFOSR Sponsored Research


1. R.S. Minns, M.R. Kutteruf, M.A. Commisso, and R.R. Jones, "Decoherence Suppression in a Resonant Driving Field," Journal of Physics B 41, 074012 (2008).

2. J. Murray-Krezan, "The Classical Dynamics of Rydberg Stark Atoms in Momentum Space," Am. J. Phys. 76 (2008).

3. X. Xhang, R.S. Minns, M.R. Kutteruf, and R.R. Jones, "Quantum Control of Rydberg Wavepackets in One- and Two-Electron Atoms," Coherence and Quantum Optics IX, N.P Bigelow, J.H. Eberly, C.R. Stroud, Jr. ed., Optical Society of America (2008).

4. M.R. Kutteruf and R.R. Jones, "Controlled Dipole-Dipole Interactions Between K Rydberg Atoms in a Laser-Chopped Effusive Beam," Physical Review A **82**, 063409 (2010).

5. M.R. Kutteruf, "Coherence in Rydberg Atoms: Measurement and Control," Ph.D. Dissertation, University of Virginia, Charlottesville, VA (2010).